

REMARKS

Status of Application

Claims 1-10 and 13-15 are pending. Claims 11-12 and 16-105 have been withdrawn as directed to non-elected inventions. These claims have been cancelled without prejudice to their prosecution in other application(s).

Claim Rejections - 35 U.S.C. § 103 (a)

Claims 1-2, 5-8, and 10 stand rejected under 35 U.S.C. § 103 (a) as being unpatentable over US 5,929,259 by Lockemeyer, in view of US Patent No. 4,118,588 by Fouquet et al. and US Patent No. 4,994,589 by Notermann.

Applicants respectfully traverse the rejection and request reconsideration of the patentability of the claims. Applicants will first address the teachings of each of the references as a whole, and then address their combination.

First, taking the teachings of Lockemeyer as a whole, Lockemeyer is directed to a method of imparting improved physical properties such as crush strength and abrasion resistance to a pre-formed **carrier** such as alpha-alumina. Lockemeyer states that adding a titania component into the mixture fired to produce the carrier results in improved crush strength and abrasion resistance, but also affects the densification of the carrier structure and that this problem increases with increasing concentrations of added titania. (Col. 1, lines 55-59) Lockemeyer concerns finding a way to incorporate the titania component into the carrier without causing such densification. Lockemeyer teaches that the way to solve the problem is to impregnate the preformed carrier with a titania generator in a liquid medium, as titania itself is not soluble in water. (Col. 2, lines 62- 63) Lockemeyer defines the term "titania generator" to "embrace all such suitable soluble titanium compounds, slurries and sols that, under the conditions under which the carrier is produced, form titania". (Col. 3, lines 9-12) Lockemeyer teaches that the calcination of the impregnated carrier is carried out under conditions adapted to generate titania. Furthermore, "[i]n the presence of alumina, the calcination can result in the formation of aluminum titanate which is, in general, less preferred than titania." (Col. 3, lines 46-50)(emphasis added)

Fouquet et al. addresses modifications to **catalysts, not to carriers**. Fouquet teaches the catalysts to be modified are "phosphates and/or silicates of magnesium, calcium, aluminum, zirconium, thorium and/or titanium, by themselves or together with oxides of magnesium, calcium, zirconium, aluminum, thorium and/or titanium and/or boric acid and/or urea". Col. 3, lines 38-42. Fouquet then uses an entire column of the patent to identify

preferred catalysts for modification by its method. (Col. 3, lines 44-68 -- Col. 4, lines 1-44)
Silver is not mentioned even once.

Fouquet also teaches that catalysts which are modified with **alkali metal carboxylates and/or alkaline earth metal carboxylates and/or alkali metal compounds and/or alkaline earth metal compounds which form carboxylates under the reaction conditions** particularly advantage the reaction. Fouquet sets forth categories of catalytic modifiers which form carboxylates, including: the carboxylates, oxides and hydroxides of lithium, sodium, potassium, magnesium and calcium, as well as of rubidium, cesium, beryllium, strontium and barium, and also alkali metal and/or alkaline earth metal silicates, borates, carbonates, bicarbonates and/or alcoholates. Fouquet also identifies as "other examples of suitable modifiers": calcium oxide and magnesium oxide, potassium magnesium silicate, calcium borate, potassium bicarbonate, lithium carbonate, sodium carbonate, sodium silicate, magnesium bicarbonate, sodium methylate, calcium ethylate and potassium propanolate. (Col. 5, lines 22-27).

Notermann teaches a process for the epoxidation of an alkene including a supported silver catalyst. As set forth in claim 1, the catalyst support contains alpha-alumina, and at least 50 per cent of the support particles having particle size of at least 0.1 micron have a platelet type morphology. Notermann **also** teaches that improved results have been observed when the support contains leachable sodium levels less than about 50 parts per million (ppm) by weight, preferably less than 40 ppm, and most preferably less than about 20 ppm based on the weight of the total support. Further, Notermann teaches that supports for use in his invention have less than a catalyst-deactivating amount of total sodium. This corresponds, in some instances, to a total sodium concentration of about 200 ppm. (Col. 13, lines 50-64)

The Examiner acknowledges that "Lockemeyer is silent with regards to the modifier being selected from among alkali and alkaline earth metal silicates", and further that Lockemeyer is silent with regards to "shaping into a lamellate or platelet morphology".

Applicant submits that Lockemeyer itself, even before combination with Fouquet et al. and Notermann, teaches away from using an aqueous solution of at least one alkali metal silicate or alkaline metal silicate as the modifier, because Lockemeyer teaches that the reaction of the modifier and the alumina carrier is **not** desirable. In contrast, the present invention is specifically directed to the reaction of the modifier with a surface of the pre-formed alpha-alumina carrier. That is, the processes of the present invention use modifiers to react with, that is, to modify the **chemical** nature of the alpha-alumina surface, in addition to physical properties, if any, which may be affected. The present application teaches:

Further, while not to be constrained by any particular theory, it is believed that during the calcination step of the present invention that the at least one alkali metal silicate and/or at least one alkaline earth metal silicate can react with the alumina surfaces, particularly in instances where there was a relatively high concentration (for example, 2 wt%) of the modifier (that is, at least one alkali metal silicate and/or at least one alkaline earth metal silicate) in the modifier impregnating solution. In the case where alpha-alumina is impregnated with sodium silicate modifier, such reaction is believed to result in the emergence of Na-Al-Si-O compounds in the alumina, for example nepheline ($\text{NaAlSi}_3\text{O}_8$). The presence of nepheline is believed to signify that a reaction as referred to above in this paragraph has occurred, however, it has been found that good performance (that is activity, efficiency and aging) is obtained whether or not nepheline is present in the modified carrier. If the temperature during the calcining carried out subsequent to the impregnation of alpha-alumina with sodium silicate is about 1400 degrees C, there is also formed carnegieite phase.

Application at p. 9, lines 29-36 through p. 10, lines 1-5.

Applicant submits that Fouquet is non-analogous art. It is directed to improving the catalyst for the manufacture of methacrylic acid and methyl methacrylate by the reaction of propionic acid and methyl propionate with dimethoxymethane. Such a reaction is far removed from the reaction in which the carrier of the present invention is to be used. As explained below with regard to Mikawa, et al., those of skill in the art are well aware that what improves a catalyst in one reaction is entirely different from what improves a catalyst in another reaction, as well as aware that a desired combination of carrier and catalyst and promoters for one reaction can be entirely different from the combination of the carrier, the catalyst, and the promoters used in a different reaction.

Second, Fouquet et al. is directed to a modification of a **catalyst, not a carrier**, using one of many possible modifiers. Carriers, much less carriers for silver-based alkene epoxidation catalysts, are **not** the subject of the modifications of Fouquet et al. One of skill in the art would not be motivated to combine the teachings of Fouquet et al. with the teachings of Lockemeyer as their respective teachings are directed to improvements in two different aspects of a catalyst: the carrier, and the catalyst. There is nothing in either Lockemeyer or Fouquet et al. that teaches that the modification of a surface of an alpha-alumina carrier is in any way desirable.

Moreover, Fouquet et al. teaches that alkali metal and/or alkaline earth metal silicates are on a laundry list of catalyst modifiers **which form carboxylates under the reaction conditions**. The carrier and catalyst of the present invention do not use modifiers which are or which form carboxylates during the reaction. Even if one of skill in the art would look to Fouquet et al, which Applicants deny, there would be no motivation to select or try any carboxylate forming modifiers, much less alkali or alkaline earth metal silicates.

In addition, given Notermann's caution regarding the presence of sodium, one of skill in the art would not be motivated to impregnate a carrier with sodium silicate unless one could be sure that such impregnation would not result in unacceptable leachable sodium levels or catalyst deactivating amounts of total sodium. Nepheline is known to be soluble under acidic conditions: See *Geochimica et Cosmochimica Acta*, Vol. 50, pp. 379-392, p. 390 (1986) (copy included), which teaches that in acid solutions, the rate of nepheline dissolution is directly proportional to the proton concentration. Notermann et al, 4,994,587 ("Notermann 2") (col. 14, lines 60-65) incorporates by reference the application which became Notermann 4,994,589. Notermann 2 teaches that leachable sodium can be determined by the treatment of the support material with a mineral acid, particularly nitric acid in a concentration of about ten percent, by volume, at a temperature of about 90 degrees C for a period of about 1 hour. (Notermann 2 at Col. 13, lines 62-68 through Col. 14, lines 1-5). A 10% nitric acid solution has a corresponding pH of below 1. A person of skill in the art would not expect to be able to incorporate nepheline into the low sodium, platelet containing support of Notermann or Notermann 2.

Claims 2-3 and 15 are rejected under 35 U.S.C. § 103 (a) as being unpatentable over US 5,929,259 by Lockemeyer, in view of US Patent No. 4,118,588 by Fouquet et al. and US Patent No. 4,994,589 by Notermann as applied to claims 1, 5-8, and 10 above, and further in view of EP 1086743 by Mikawa et al.

Applicants respectfully traverse this rejection. Applicants' comments regarding Lockemeyer, Fouquet et al. and Notermann are set forth above. The Examiner states that Mikawa teaches a method of making a catalyst for the production of epoxides by a vapor-phase oxidation of an unsaturated hydrocarbon, wherein a sodium silicate modifier along with an alpha-alumina carrier is used (citing claim 8) and that the modifier may be a sodium silicate with a stoichiometry of $\text{Na}_2\text{O} \cdot 2.6\text{SiO}_2$ (citing claim 4). Applicant notes that claim 8 of Mikawa does not refer to sodium silicate and that claim 4 of Mikawa does not reference a sodium silicate of any particular stoichiometry. In [0034], a number of silicon compounds are mentioned: silicon oxide, silicon nitride, silicon carbide, silane, and silicon sulfate, silicates such as sodium silicate, ammonium silicate, sodium aluminosilicate, ammonium aluminosilicate, sodium phosphosilicate, and ammonium phosphosilicate, complex salts of silica containing such silicon as feldspar and clay, and silica mixture.

Moreover, Mikawa is non-analogous art, as one of skill in the art of making a carrier to be used in the vapor phase epoxidation of ethylene would not look to art concerning the oxidation of unsaturated hydrocarbons **having 4 to 20 carbon atoms** (Mikawa, paragraph

[0001]). Mikawa itself teaches that the catalysts which are used for the production of ethylene oxide, when used in the reaction for producing 3, 4-epoxy-1-butene by the catalytic vapor phase oxidation of 1, 3-butadiene **“generally manifest substantially no catalytic activity, or if catalytically active at all, offer very short service lives as a catalyst”**. (Mikawa, paragraph [0004])

Other references also teach that catalysts used in the epoxidation of ethylene do not have the same effects as catalysts for the epoxidation of olefins having a higher number of carbon atoms. See, e.g., Monnier, The role of Alkali Promoters in the Ag-catalyzed Epoxidation of Olefins, 234th ACS National Meeting, Boston, MA, United States, August 19-23, 2007 (2007) (cesium promoted silver based catalysts used in epoxidation of higher olefins have higher activity, selectivity, and extended catalyst lifetimes, whereas cesium promoted silver based catalyst used to epoxidate ethylene oxide increase selectivity at the expense of activity; rubidium and titanium promoters exhibit similar behavior similar to cesium for butadiene epoxidation, while potassium promoters have no effect on Ag for butadiene epoxidation); Monnier, et al., Stability and Distribution of Cesium in Cesium Promoted Silver Catalysts used for Butadiene Epoxidation, Journal of Catalysis (2004), 226(2), 401-409 (catalysts used for butadiene epoxidation contain much higher levels of cesium promoters (600-1400 ppm) compared to catalysts used for ethylene epoxidation (200-400 ppm)); Monnier, et al., The Direct Epoxidation of Higher Olefins using Molecular Oxygen, Applied Catalysis, A: General (2001), 221(1-2), 73-91 (optimum catalyst formulations for propylene or C4 epoxidation are different from those for ethylene epoxidation) .

In addition, the combination of Mikawa et al. with the other three references does not render the claims obvious to one of skill in the art at the time the invention was made. Mikawa, et al., describes in great detail the method of **making the carrier**, by mixing an alpha alumina **powder** with a particular sodium content with an aluminum compound, a silicon compound and a sodium compound, followed by calcination of the mixture. Mikawa, et al. does not describe or suggest treating a **preformed carrier** with any modifier, let alone an alkali metal or alkaline earth metal silicate modifier.

Furthermore, given that Mikawa et al. describes a number of possible combinations of sodium compounds and a number of possible silicon compounds to add to the carrier precursor, the selection by the Applicants of an alkali metal silicate or alkaline earth metal silicate to use as a modifier—particular post carrier formation—is not obvious.

Additionally, if Mikawa et al. is cited to provide the motivation to combine sodium silicates as a modifier with Lockemeyer's post-treatment methodology, Mikawa et al. teaches

that the roles of the added silicon and sodium compounds are quite different. Paragraph [0026] indicates that the added aluminum and silicon compounds desirably form $Al_6Si_2O_{13}$, which is believed to influence the acidity of the surface of the carrier. This is further supported by the teaching in paragraph [0018] of Mikawa et al. that if the silicon content is below certain limits, "the amount of acid on the surface of the carrier will be unduly small and the effect due to the acidity of the surface will manifest with difficulty." In contrast, on the role of sodium, consistent with paragraph [0022], paragraph [0025] of Mikawa et al. teaches that "if the sodium content is unduly low, the strength of the carrier will be degraded. The sodium content may depend on the amount of silicon in the carrier. If the sodium content is unduly large, the surface acidity will be lost..." In other words, silicon is desired for a **chemical** improvement via provision of surface acidity whereas sodium improves **physical** strength even while undesirably counteracting the acidity provided via silicon. Given these opposing effects, a skilled person would clearly prefer to be able to vary the levels of sodium and silicon independently in order to balance and optimize both sets of effects instead of being constrained to certain $SiO_2:Na_2O$ ratios as would be provided by use of sodium silicate compounds of particular stoichiometry. This is further supported by the generally high $SiO_2:Na_2O$ ratios (relative to nepheline) in the fired carriers of Mikawa's examples, as well as the notable non-usage of sodium silicates in preparing these carriers despite the reference in paragraph [0034].

Assuming only for the purposes of this argument that either Fouquet or Mikawa are proper references, which Applicants do not concede, using the modifier of Mikawa in the "modified process of Lockemeyer" does **not** merely "provide a functional equivalent":

(1) Lockemeyer expressly teaches how to make a preformed **carrier** with improved **physical characteristics** because the performed carrier was impregnated with a titania modifier. It counsels that a chemical reaction between the modifier and alumina results is undesirable.

(2) Fouquet teaches how to modify certain metal **catalysts**, preferentially by using carboxylate or carboxylate generating modifiers, which are selected from a laundry list that includes alkali metal silicates and alkaline earth metal silicates.

(3) Mikawa teaches a method of making a carrier made by adding any one of a number of sodium and any one of a number of silicon compounds to **carrier precursor** materials.

Only with the benefit of impermissible hindsight, that is, having the benefit of the teachings of the present application, would one of skill in the art of making ethylene oxide carriers be motivated to impregnate an alkali metal or alkaline earth metal silicate modifier

on a **pre-formed alpha-alumina carrier followed by calcination wherein a surface of the alpha-alumina is reacted with the modifier.**

The remaining rejections: With regard to claim 14, please see applicants' response regarding the rejection of claims 2-3 above. In addition, for the reasons set forth above, the independent claims are patentable, and the rejections to the dependent claims are moot.

Applicants respectfully requests reconsideration of the rejection of claims 1-10 and 13-15 and further submit that claims 1-10 and 13-15 are in condition for allowance.

Respectfully submitted,

/Lois K. Ruzala/Reg No 39074/

Lois K. Ruzala
Registration No. 39,074
Phone: (989) 636-1556

P. O. Box 1967
Midland, MI 48641-1967

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